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SURFACE REACTIONS AND FATIGUE CRACK GROWTH

by

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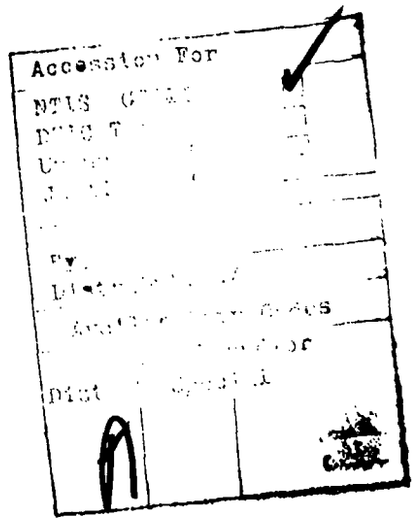

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INTRODUCTION

Recent fracture mechanics and surface chemistry studies of environment assisted crack growth in gaseous environments[1-4] have shown that crack growth may be controlled in some systems by the rate of surface reactions and in others by the rate of transport of the aggressive environment to the crack tip. Based on considerations of surface reactions and gas transport, a model for surface reaction and transport controlled fatigue crack growth in single component gaseous environments was proposed and experimentally verified.[5-8] This model is able to account for the influences of gas pressure and cyclic load frequency on the rate of fatigue crack growth. In practice, however, there is usually more than one gas in a given environment. The various component gases can compete for surface reaction sites and therefore alter the fatigue crack growth response. The effect of this competition needs to be considered. In this paper, the development of the model for surface reaction and transport controlled fatigue crack growth in single component gaseous environments is briefly reviewed. Extension of the same considerations to fatigue crack growth in a binary gas mixture, in which one of the components acts as an inhibitor, is described. Quantitative application of this model to the consideration of oxygen on fatigue crack growth in humid air is discussed.

MODELING OF FATIGUE CRACK GROWTH IN ONE COMPONENT GAS

Modeling of environment assisted fatigue crack growth in single-component gaseous environments was based on the proposition that the rate of crack growth in an aggressive environment, $(da/dN)_e$, is

composed of the sum of three components.[5,8,10]

$$(da/dN)_e = (da/dN)_r + (da/dN)_{cf} + (da/dN)_{scc} \quad (1)$$

$(da/dN)_r$ is the rate of fatigue crack growth in an inert environment and, therefore, represents the contribution of "pure" (mechanical) fatigue. This component is essentially independent of frequency at temperatures where creep is not important. $(da/dN)_{scc}$ is the contribution by sustained-load crack growth (that is, stress corrosion cracking) at K levels above K_{Iscc}.[11] The contribution by sustained-load crack growth, *i.e.*, the $(da/dN)_{scc}$ term, has been examined in some detail previously,[11,12] and appears to be adequately accounted for by the superposition model.[11] $(da/dN)_{cf}$ represents a cycle-dependent contribution requiring synergistic interaction of fatigue and environmental attack, and was not considered by Wei and Landes.[11] This model,[5] therefore, was directed specifically to the cycle-dependent term, and was limited to establishing a formal framework for estimating frequency and pressure dependence of $(da/dN)_{cf}$ in single-component gaseous environments. (Examinations of the influences of other loading variables (such as, load ratio) are in progress.)

In the model,[5] environmental enhancement of fatigue crack growth is assumed to result from embrittlement by hydrogen that is produced by the reaction of hydrogenous gases (*e.g.*, water vapor) with the freshly produced fatigue crack surfaces. More specifically, $(da/dN)_{cf}$ is assumed to be proportional to the amount of hydrogen produced by the surface reaction during each cycle, which is proportional in turn to the "effective" crack area[1/] produced during the prior loading cycles and to the extent of surface reaction. The time available for reaction is assumed to be equal to one-half of the fatigue cycle.[5] Transport of gaseous environments to the crack tip is assumed to be by Knudsen flow[13] at low pressures. The rates of hydrogen diffusion and embrittlement were assumed to be much faster than those of gas transport and surface reaction, and, therefore, did not need to be considered.

The governing differential equations for flow and surface reactions, and the relationship for $(da/dN)_{cf}$ are as follows:[5]

[1/] In the original derivation,[5] it was assumed that only the area produced by the previous cycle of crack growth contributed to embrittlement. It is recognized that a greater portion of the crack surface remained active, particularly for slow surface reactions and at high frequencies, and can contribute hydrogen to the embrittlement process.[8] To provide consistency, while retaining the form of the original model, an "effective" crack area or "effective" crack increment Δa^* is defined here.

$$\frac{dp}{dt} = -\frac{SN_oRT}{V}\frac{d\theta}{dt} + \frac{F}{V}(p_o - p) \qquad (2)$$

$$\frac{d\theta}{dt} = k_c p f(\theta) = k_c p(1 - \theta) \qquad (3)$$

$$(da/dN)_{cf} \propto \theta \cdot \Delta a^{*}\text{[1]} \qquad (4)$$

The terms in the equations are as follows:[5]

F = $8.72 \times 10^2\ \beta(\sigma_{ys}/E)^2\ (T/M)^{1/2}\ B\ell$ (in m^3/s) = Knudsen flow parameter that depends on dimension and shape of the capillary, molecular weight (M) of the gas and temperature (T). The specific form of this expression reflects an attempt to account for constriction in flow by the real crack, where ℓ is a selected distance (of the order 10^{-6} m) from the crack tip used in defining a crack opening and β is an empirical quantity to be determined from the crack growth data.[5,13]

k_c = reaction rate constant; $(Pa\text{-}s)^{-1}$.

N_o = density of surface sites; molecules (atoms)-m^{-2}.

p = pressure of gas at the crack tip; Pa.

p_o = pressure of gas in the surrounding environment; Pa.

R = gas constant = 1.38×10^{-23} Pa-m^3-molecules^{-1} - K^{-1}.

S = area of "effective" crack surface per cycle = $\alpha\ (2B\Delta a^*)$, where Δa^* = "effective" increment per cycle, B = specimen thickness, and α = empirical constant for surface roughness and crack geometry.[5]

T = absolute temperature.

V = control volume at the crack tip, *i.e.*, volume associated with the distance ℓ.

θ = fractional surface coverage or extent of reaction of surface per unit area.

From eqn. (2), it can be seen that the rate of change of pressure at the crack tip depends on the decrease in pressure produced by reaction of the environment with the active ("effective") crack surface and on the increase in pressure from the influx of gas from the external environment. The form $f(\theta) = 1 - \theta$ in eqn. (3) incorporates the assumption that the surface reaction is first-order in relation to available surface sites.[4-7]

Solutions for eqns. (2) and (3), with $f(\theta) = 1 - \theta$, were obtained for two limiting cases, for $0 < \theta < 1$, and were used in

conjunction with eqn. (4) to estimate the influences of key environmental and loading variables on the cycle-dependent component of fatigue crack growth rate, *i.e.*, on $(da/dN)_{cf}$.[5]

Case I: Transport controlled.

$$\theta \approx \frac{F}{SN_oRT} p_o t \quad \text{for} \quad \frac{SN_oRTk_c}{F} >> 1 \tag{5}$$

Case II: Surface reaction controlled.

$$\theta \approx 1 - \exp(-k_c p_o t) \quad \text{for} \quad \frac{SN_oRTk_c}{F} << 1 \tag{6}$$

In the transport controlled case, because of the rapid reactions of the environment with the freshly created crack surfaces (high k_c) and the limited rate of supply of the environment to the crack tip, significant attenuation of gas pressure takes place at the crack tip. The extent of surface reaction (θ) during one cycle is controlled by the rate of transport of the aggressive environment to the crack tip, and thus varies linearly with time (see eqn. (5)). For the surface reaction controlled case, the reaction rates are sufficiently slow so that the gas pressure at the crack tip is essentially equal to the external pressure. The extent of reaction, for $f(\theta) = 1 - \theta$, becomes an exponential function of time, eqn. (6).

As a modification to the original model,[5,8] the "effective" crack increment (Δa^*) is now chosen, along with ℓ in the expression of F, to be equal to the growth increment per cycle at saturation (*i.e.*, for $\theta \approx 1$, $\Delta a^* = \ell = (da/dN)_{e,s} \cdot 1$). By taking t equal to $\tau/2$ or $1/2f$,[5,8] the following expressions can be obtained from eqns. (4), (5) and (6):

Case I: Transport controlled.

$$(p_o/2f)_s \approx \frac{SN_oRT}{F} = \left[4.36 \times 10^2 \frac{\beta}{\alpha} \frac{\sigma_{ys}^2}{N_oRTE^2}(T/M)^{1/2}\right]^{-1} \tag{7}$$

$$\frac{(da/dN)_{cf}}{(da/dN)_{e,s}} \approx 4.36 \times 10^2 \frac{\beta}{\alpha} \frac{\sigma_{ys}^2}{N_oRTE^2}(T/M)^{1/2} \frac{p_o}{2f} \tag{8}$$

$$\frac{(da/dN)_{cf}}{(da/dN)_{cf,s}} = \frac{(p_o/2f)}{(p_o/2f)_s} \tag{9}$$

Case II: Surface reaction controlled.

$$\frac{(da/dN)_{cf}}{(da/dN)_{e,s}} \propto 1 - \exp(-k_c p_o/2f) \tag{10}$$

$$\frac{(da/dN)_{cf}}{(da/dN)_{cf,s}} = 1 - \exp(-k_c p_o/2f) \tag{11}$$

Subscript s is used to denote the corresponding values at saturation. The modification affects only the form of the expressions for the surface reaction controlled case, and provides a physically more acceptable model. In their present form, the model appears to be in excellent agreement with the experimental observations[2/].[5-9,14] An example of this correlation is illustrated in Fig. 1 for transport controlled crack growth in a 2219-T851 aluminum alloy.[6]

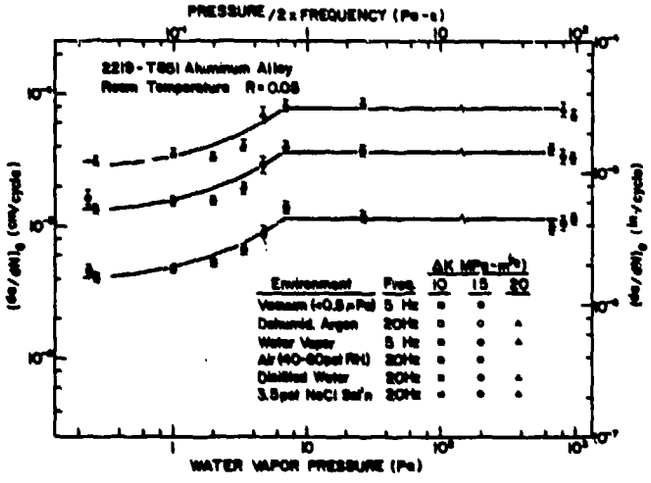

Fig. 1. Influence of water vapor pressure (or pressure/2xfrequency) on fatigue crack growth rates in 2219-T851 aluminum alloy at room temperature. Solid lines represent model predictions.[5,6]

2/ There is a discrepancy of 10^2 in exposure ($p_o/2f$) between the model predictions and the experimental data on AISI 4340 steel,[9] suggesting that capillary condensation of water vapor may have taken place. With capillary condensation, the effective exposure can be much higher than that given by $p_o/2f$.

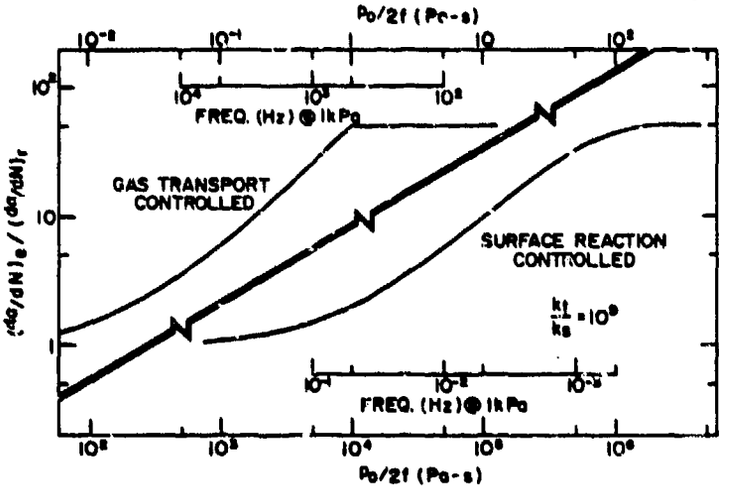

Fig. 2. Schematic illustration and comparison of gas transport and surface reaction controlled fatigue crack growth.

The engineering significance and implications of the model may be discussed in relation to eqns. (7) and (11), and considered through the schematic illustration shown in Fig. 2. The model provides a formalism for "predicting" environmentally assisted fatigue crack growth response in relation to gas pressure and cyclic load frequency. Specifically, eqn. (7) gives, for the transport controlled case, the value of $p_o/2f$ at which "saturation" in environmental effect can be expected in terms of the properties of the alloy, molecular weight of the aggressive gas, and temperature. Fatigue crack growth response is given, for this case, as an explicit function of $p_o/2f$ below saturation by eqns. (8) and (9). For the surface reaction controlled case, the dependence of fatigue crack growth response on $p_o/2f$ is given by eqns. (10) and (11). It should be noted that eqns. (8) to (11) deal only with relative rates. The actual growth rates depend on the interactions of the embrittling specie (hydrogen) with the alloy, which are not adequately understood at this time.

As further illustration, two hypothetical cases -- one representing gas transport and the other surface reaction control -- are shown in Fig. 2. Crack growth response curves are given in terms of the ratio $(da/dN)_e/(da/dN)_r$, as functions of $p_o/2f$, which can be readily obtained from eqns. (1), (8) and (11). In addition, frequency scales are shown for an external pressure (p_o) of 1 kPa. For this illustration, $(da/dN)_{e,s}/(da/dN)_r = 50$, and $k_c = 2 \times 10^3$ and 2×10^{-6} $(Pa\text{-}s)^{-1}$ are used. Although the curves differ somewhat in detail from experimental data, they do serve to illustrate fatigue crack growth response for a high strength steel exposed to hydrogen

sulfide (transport controlled) and water vapor (reaction controlled).[7] It can be seen that the range of exposures ($p_o/2f$) or frequencies over which apparent frequency dependence may be observed differ by about 6 orders of magnitude for the two cases. The apparent independence of frequency or pressure, or $p_o/2f$, does not imply absence of environmental effect. Neither does testing at high frequencies in itself ensure absence of environmental effects. Most importantly, each material-environment system must be considered individually. Generalizations and extrapolations on the basis of limited data can be very misleading and should be avoided.

MODELING OF FATIGUE CRACK GROWTH IN BINARY GAS MIXTURE

Experimental evidence obtained to date[1-4,6-9] clearly indicates that the rate of environment assisted crack growth is dependent on the rate of hydrogen production at the crack surfaces. The model for fatigue crack growth developed earlier[5,8] may be readily extended to the consideration of crack growth in gas mixtures. For simplicity, the case of a binary mixture is considered. One of the component gases is taken to be an inhibitor (that is, a gas which will react with the clean metal surface but will not enhance crack growth). As such, the results may be used, for example, to examine the influence of oxygen (an inhibitor) on fatigue crack growth in humid air, where water vapor is the aggressive component.[14-16]

It is assumed that (i) both gases are strongly adsorbed on the clean metal surface, (ii) chemical adsorption of either gas at a given surface site precludes further adsorption at that site, (iii) the ratio of partial pressures of the gases at the crack tip is essentially the same as that of the surrounding environment, and (iv) no capillary condensation of either gas occurs at the crack tip. In line with the model of Weir et al.,[5] the cycle-dependent component of crack growth rate in the gas mixture, $(da/dN)_{cf,m}$, is assumed to be proportional to the extent of surface reaction with the aggressive gas during one loading cycle, or to θ_a. If one now assumes, for simplicity, that the kinetics of surface reaction for both gases are first order with respect to pressure and to available surface sites, then the rate equations for the surface reactions may be written as follows:

$$\frac{d\theta_a}{dt} = k_a p_a (1 - \theta) \tag{12}$$

$$\frac{d\theta_i}{dt} = k_i p_i (1 - \theta) \tag{13}$$

where the subscripts a and i denote the aggressive and inhibitor gases respectively. The quantities k_a, p_a, k_i and p_i are the

reaction rate constants and partial pressures of the gases at the crack tip, respectively. The coverages θ_a and θ_i denote the fraction of surface that has reacted with the aggressive and inhibitor gases, respectively. Coverage for both gases is denoted by θ, where $\theta = \theta_a + \theta_i$ and $0 \leq \theta \leq 1$.

Eqns. (12) and (13) may be solved straightforwardly to obtain the extent of reaction of a fresh surface with each gas after being exposed to the gas mixture for a time t.

$$\theta_a = \frac{k_a p_a}{k_a p_a + k_i p_i}\{1 - \exp[-(k_a p_a + k_i p_i)t]\} \tag{14}$$

$$\theta_i = \frac{k_i p_i}{k_a p_a + k_i p_i}\{1 - \exp[-(k_a p_a + k_i p_i)t]\} \tag{15}$$

It follows that the fraction of surface that has reacted with the aggressive gas (θ_a) in relation to the total surface coverage (θ) may be given by eqn. (16).

$$\frac{\theta_a}{\theta} = \frac{\theta_a}{\theta_a + \theta_i} = \left(1 + \frac{k_i p_i}{k_a p_a}\right)^{-1} \tag{16}$$

If the total pressure of the gas mixture at the crack tip ($p_m = p_a + p_i$) and cyclic-load frequency, or $p_m/2f$, are such to produce "complete" surface reaction or "saturation" coverage ($\theta \approx 1$) during one cycle, then the fraction of the crack surface that has reacted with the aggressive gas is given by eqn. (17).

$$\theta_a = \left(1 + \frac{k_i p_i}{k_a p_a}\right)^{-1} \quad ; \quad (\theta \approx 1) \tag{17}$$

Since $(da/dN)_{cf} \propto \theta \cdot \Delta a^*$ (see eqn. (4)), the cycle-dependent component of fatigue crack growth rate in the mixed gases, $(da/dN)_{cf,m}$, can be obtained from the cycle-dependent rate in a pure gas at saturation, $(da/dN)_{cf,p,s}$, and eqn. (17).

$$\frac{(da/dN)_{cf,m}}{(da/dN)_{cf,p,s}} = \left(1 + \frac{k_i p_i}{k_a p_a}\right)^{-1} = \left(1 + \frac{k_i p_{io}}{k_a p_{ao}}\right)^{-1} \tag{18}$$

(for $\theta \approx 1$)

The subscript o is used to denote pressures in the external environment, and reflects the assumption that the partial pressure ratio at the crack tip is the same as that in the external environment. It should be noted that only the competition between two gases has been modeled here, and eqn. (18) is expected to apply equally well to both transport and surface reaction controlled crack growth. Schematic illustration of eqn. (18) is shown in Fig. 3 as $(da/dN)_{e,m}/(da/dN)_r$ for different ratios of $(da/dN)_{e,s}/(da/dN)_r$; the ratios reflect different degrees of severity of environmental effects in the pure gas. A ratio of k_i/k_a of 0.14, which approximates the ratio between the reaction rate constants for oxygen and water vapor over a 2219-T851 aluminum alloy,[6,17] is used. The upper scale for p_a in Fig. 3 is computed for p_i = 152 torr (or 20.2 kPa), and therefore corresponds to the partial pressure of water vapor in humid (moist) air.

Assuming that the surface reaction data on 2219-T851 aluminum alloys[6,17] may be used for other high-strength aluminum alloys, comparisons of model predictions with data reported by Hartman et al.[15] and Feeney, McMillan and Wei[16] on 7075-T6 aluminum alloy, and Bradshaw and Wheeler[14] on DTD 5070A aluminum alloy are made and are shown in Figs. 4 and 5, respectively. For the 7075-T6 aluminum alloy, data in water and salt water[16] are assumed to represent the "saturation" level in pure water vapor (θ = 1).[6] Excellent agreement is observed at the higher K levels and higher crack growth rates. At the lower K levels and lower crack growth rates, the data tend to follow those of pure water vapor, and may reflect partial capillary condensation during a portion of each loading-unloading

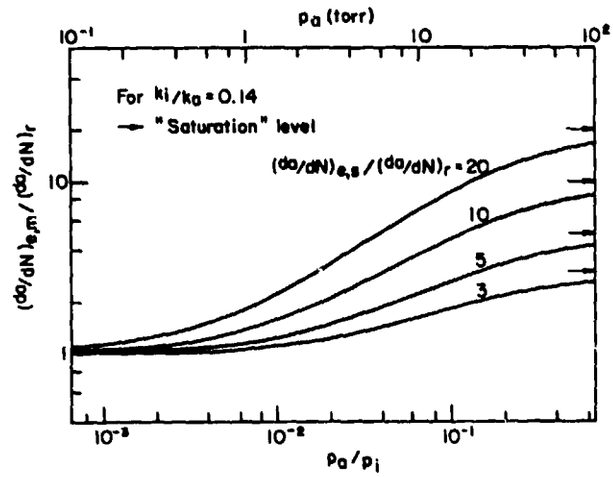

Fig. 3. Schematic illustration of the influence of inhibitor gas on fatigue crack growth in a binary gas mixture. (In the upper scale, p_a corresponds to p_i = 152 torr or 20.2 kPa.)

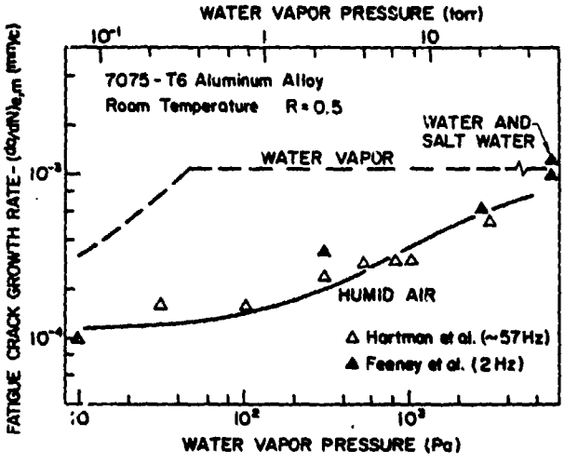

Fig. 4. Influence of water vapor pressure on fatigue crack growth in 7075-T6 aluminum alloy in humid air.[15,16] Dashed and solid lines represent model predictions from eqns. (7), (8) and (18).

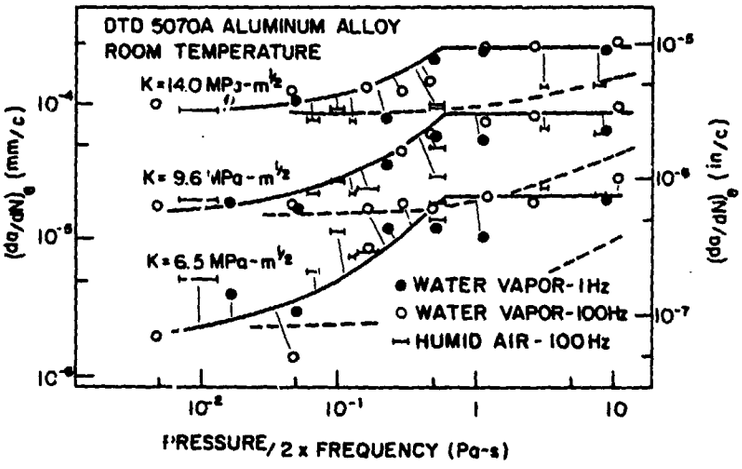

Fig. 5. Influence of water vapor pressure and frequency on fatigue crack growth in DTD 5070A aluminum alloy.[14] Solid lines represent model predictions for water vapor, and dashed lines for humid air.

cycle or some other factor. Further studies are needed to clarify this discrepancy.

DISCUSSION

On the basis of available experimental results, it is seen that the models reasonably account for environmentally assisted fatigue crack growth response in single-component gaseous environments and in binary gas mixtures, in which one of the components acts as an inhibitor. A similar consideration of environmentally assisted crack growth in binary gas mixtures, under sustained loading, has also been made and provided good agreement with experimental observations.[18] Thus, the models provide a formalism for the planning of experiments, and for the interpretation and extrapolation of data for engineering applications.

In deriving these models and in their application, only simple surface reactions have been considered. Many reactions that are of interest (for example, reactions of steels with water vapor or hydrogen sulfide), however, are more complex. Reactions of different components to form new complexes are also possible. Incorporation of these complexities into future modeling efforts must be considered. Extension of these ideas to the consideration of fatigue crack growth in mixtures of H_2S and CO has been made.[19,20] Preliminary results indicate that modeling of the more complex environments would be feasible.

SUMMARY

Based on considerations of surface reactions and gas transport, models of environmentally assisted fatigue crack growth in single-component gaseous environments and in binary gas mixtures (in which one of the components acts as an inhibitor) have been developed. Good agreement with experimental data has been observed. The models, therefore, provide a formalized framework for experimental design, and for the interpretation and extrapolation of data for engineering applications. Further research will be needed to refine and extend this work to the consideration of more complex environments, and to incorporate the influences of metallurgical variables.

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